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# Statistical Mechanics of Non-Equilibrium Systems : Extensive Property, Fluctuation and Nonlinear Response

AUTHOR(S):

SUZUKI, Masuo

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Statistical Mechanics of  
Non-Equilibrium Systems

— Extensive Property, Fluctuation and Nonlinear Response —

Masuo SUZUKI (Dept. of Physics, Univ. of Tokyo)

(to be submitted to Prog. Theor. Phys.)

1. Kubo's Ansatz: Extensive Property

$$P(X, t) = C \exp [\Omega \phi(x, t)] ,$$

for large  $\Omega$  with  $x = X/\Omega$ ;  $X$  = macrovariable.

2. When  $X$  is a Markoffian macrovariable,

$$\frac{\partial P(x, t)}{\partial t} = \Gamma_M P(x, t) ; \Gamma_M = \text{time develop. op.}$$

evol. eq. for  $y(t)$  = deterministic path :

$$\dot{y}(t) = C_1(y(t)) ; \quad C_n = n\text{-th moment},$$

variance  $\sigma(t)$  :

$$\dot{\sigma}(t) = 2 C_1'(y(t)) \sigma(t) + C_2(y(t))$$

3. When  $X$  is a non-Markoffian macrovariable,

$$\frac{\partial P(\{\sigma_j\}; t)}{\partial t} = \Gamma P(\{\sigma_j\}; t),$$

$$P(X, t) = \sum^{\text{config.}} \delta(\mathbf{X} - X) P(\{\sigma_j\}; t)$$

4. For the macrovariable  $X$  in quantum systems,

$$\rho(X, t) = T_r \delta(\mathbf{X} - X) \rho(t) ; \quad i\hbar \frac{\partial \rho(t)}{\partial t} = [\mathcal{H}, \rho(t)]$$

Extensive Property:

$$\rho(X, t) = C \exp [\Omega \phi(x, t)]$$

5. Generating Function Formalism

$$\Psi(\lambda, t) = \begin{cases} T_r e^{\lambda \mathbf{X}} \rho(t) \dots\dots\dots (\text{quantal}) \\ \sum_{\text{config.}} e^{\lambda \mathbf{X}} e^{t\Gamma} P_0 \dots\dots\dots (\text{stochastic or classical}) \end{cases}$$

$$\rho(X, t) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} e^{-\lambda X} \Psi(\lambda, t) d\lambda$$

generalized thermodynamic potential

$$\mathcal{F}(\lambda, t) \equiv \log \Psi(\lambda, t) \text{ or } \Psi(\lambda, t) = e^{\mathcal{F}(\lambda, t)}$$

6. Assumption for the initial distribution:

$$\rho(0) = e^{\mathcal{H}(i)} \text{ or } P_0 = e^{\mathcal{H}(i)} \quad (\text{canonical})(\text{normalized})$$

7. Properties of  $\Psi(\lambda, t)$  and  $\mathcal{F}(\lambda, t)$  when  $X, \mathcal{M}^{(i)}$  and  $\mathcal{M}$  are Hermitian:

- 1)  $\Psi(\lambda, t) > 0$  and  $\mathcal{F}(\lambda, t) = \text{real}$  for  $\lambda$  real.
- 2) When  $\lambda$  is real,  $\Psi$  and  $\mathcal{F}$  are convex functions of  $\lambda$ .
- 3) For complex  $\lambda$  (= not real), we have

$$|\Psi(\lambda, t)| < \Psi(\text{Re } \lambda, t)$$

8. If  $\Psi(\lambda, t) = C_1 e^{\Omega \psi(\lambda, t)}$  (Extensive property),

$$\begin{aligned} \rho(X, t) &= \frac{C_1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \exp \left[ -\Omega \{ \lambda x - \psi(\lambda, x) \} \right] d\lambda \\ &= C_2 \exp \left[ \Omega \{ \psi(\lambda_0, t) - \lambda_0 x \} \right] \end{aligned}$$

for large  $\Omega$ ,  $\lambda_0$  being given by the solution

$$\frac{\partial \psi(\lambda, t)}{\partial \lambda} = x$$

9. "Local operator"  $Q(\mathbf{r})$  is defined by a functional of field operators  $\{\psi(\mathbf{r}')\}$ ;  $\mathbf{r}' \in D(\mathbf{r}) \equiv \text{circle domain of radius } b$ .
10. General conditions for  $X, \mathcal{M}^{(i)}$  and  $\mathcal{M}$  :

$$\mathbf{X} = \int \mathbf{X}(\mathbf{r}) d\mathbf{r}, \mathcal{M}^{(i)} = \int \mathcal{M}^{(i)}(\mathbf{r}) d\mathbf{r} \text{ and } \mathcal{M} = \int \mathcal{M}(\mathbf{r}) d\mathbf{r}.$$

11. Theorem for "extensive property" in quantal systems.

Theorem I (quantal) If the averages of local operators,

$\langle \mathbf{X}(\mathbf{r}) \rangle^{(1)}$ ,  $\langle \mathcal{M}^{(i)}(\mathbf{r}) \rangle^{(2)}$ , and  $\langle \mathcal{M}(\mathbf{r}) \rangle^{(3)}$  are bounded, then

$$\lim_{\Omega \rightarrow \infty} \psi_{\Omega}(\lambda, t) = \text{exist (uniformly convergent)}$$

for  $|\lambda| < A$  (fixed) and  $t$  finite. Therefore  $\Psi(\lambda, t)$  has the

extensive property:  $\Psi(\lambda, t) = C_1 \exp [\Omega \psi(\lambda, t)]$

and consequently so does  $\rho(X, t)$ .

## 12. Definition of the averages

$$\langle \mathbf{X}(\mathbf{r}) \rangle^{(1)}, \langle \mathcal{H}^{(i)}(\mathbf{r}) \rangle^{(2)} \text{ and } \langle \mathcal{H}(\mathbf{r}) \rangle^{(3)} :$$

First separate the system  $\Omega$  into  $\Omega_1$  and  $\Omega_2$  :

$$\mathbf{X} = \mathbf{X}_1 + \mathbf{X}_2, \quad \mathcal{H}^{(i)} = \mathcal{H}_1^{(i)} + \mathcal{H}_2^{(i)} \text{ and } \mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2,$$

$$\text{where } Q_j = \int_{\Omega_j} Q(\mathbf{r}) d\mathbf{r}.$$

$$\text{Operational; } P_{(s,\mu)} Q \equiv e^{-s(P_1 + \mu P_2)} Q e^{s(P_1 + \mu P_2)},$$

$$\langle Q \rangle^{(j)} \equiv T_r Q \rho_j / T_r \rho_j,$$

$$\text{where } \rho_j = \rho_j(s, \mu), \quad 0 \leq s \leq 1, \quad 0 \leq \mu \leq 1,$$

$$\begin{cases} \rho_1(s, \mu) \equiv \exp [\lambda (X_1 + \mu X_2)] \exp [\lambda X_{(s,\mu)} \mathcal{H}_{(s,\mu)} \mathcal{H}^{(i)}], \\ \rho_2(s, \mu) \equiv \exp [\mathcal{H}_1^{(i)} + \mu \mathcal{H}_2^{(i)}] \exp [\lambda \mathcal{H}_{(s,\mu)}^{(i)} \mathcal{H}_{(-it,1)} X_1] \\ \rho_3(s, \mu) \equiv \exp [\mathcal{H}_{(ist,\mu)} \mathcal{H}_1^{(i)}] \exp [\lambda \mathcal{H}_{(i(s-1)t,\mu)} X_1] \end{cases}$$

 13. Remarks on Theorem I

a)  $\Psi$  is a continuous function of  $\lambda$ .

b) "Bounded"  $\Rightarrow |\langle Q(\mathbf{r}) \rangle^{(j)}| \leq c_j$  (indep. of  $\Omega$ ).

c) If  $\{c_j\}$  are indep. of  $\lambda$  for  $|\lambda| < A$ , the limit  $\psi_\Omega(\lambda, t) \Rightarrow$

$\psi(\lambda, t)$  is uniformly convergent in wider sense in the complex circle domain  $|\lambda| < A$ . Hence,

$$\lim_{\Omega \rightarrow \infty} \frac{d^p \psi_\Omega(\lambda, t)}{d\lambda^p} = \frac{d^p \psi(\lambda, t)}{d\lambda^p} \quad (\text{Weierstrass})$$

d) "bounded" for  $\Omega_2/\Omega_1 = O(\Omega^{-1/d}) \Rightarrow \text{O.K.}$

e) "bounded" for  $s = \mu = 1 \Rightarrow "$  for  $0 < s < 1, 0 < \mu < 1$  (conjecture)

f) When  $\mathbf{X}(\mathbf{r})$  commutes with  $\mathbf{X}(\mathbf{r}')$ ,  $\langle \mathbf{X}(\mathbf{r}) \rangle^{(1)}$  is bounded.

g) If  $[\mathcal{H}^{(i)}(\mathbf{r}), \mathcal{H}^{(i)}(\mathbf{r}')] = 0 \Rightarrow \langle \mathcal{H}^{(i)}(\mathbf{r}) \rangle^{(1)}$  is bounded.

It is not easy to prove that  $\langle \mathcal{H}(\mathbf{r}) \rangle^{(3)}$  is bounded when  $[\mathcal{H}(\mathbf{r}), \mathcal{H}(\mathbf{r}')] = 0$ .

- h) Clearly,  $\langle \mathcal{X}(\mathbf{r}) \rangle^{(3)}$  is bounded if  $\mathcal{X}_2 \equiv 0$ .
- i) Only the boundness of  $\langle \mathbf{X}(\mathbf{r}) \rangle^{(1)}$  etc. are assumed.
- j)  $\lim_{\Omega \rightarrow \infty} =$  thermodynamic limit in van Hove's sense.
- k) Theorem I is extended to classical systems. (Classical Liouville op.).

14. Proof of Extensive Property:

Separation of the domain  $\Omega$

$$\Omega_n = \Omega_1 + \Omega_2$$

$n = \text{large integer}$

$$\begin{cases} \mathbf{X} = \mathbf{X}_1 + \mathbf{X}_2 \\ \mathcal{X}^{(i)} = \mathcal{X}_1^{(i)} + \mathcal{X}_2^{(i)} \\ \mathcal{X} = \mathcal{X}_1 + \mathcal{X}_2 \end{cases}$$

To prove Cauchy's condition on convergence.

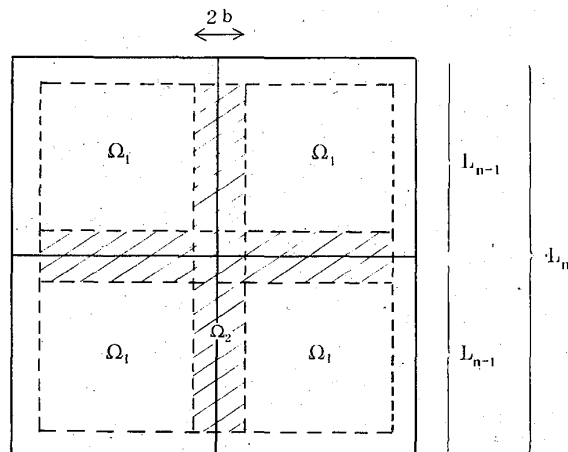


図1 Domains  $\Omega_1$  and  $\Omega_2$  with each margin of width  $b$  inside in it;  $\Omega_2$  is the shaded region.

$$|\psi_n(\lambda, t) - \psi_{n-1}(\lambda, t)| \leq 2^{-n} (Ac_1 + c_2 + 2tc_3)(2bd),$$

$d = \text{dimension},$

$$\therefore |\psi_{n+m}(\lambda, t) - \psi_n(\lambda, t)| \leq 2^{-n} ( \quad \quad \quad ),$$

Formula.  $\therefore \lim_{n \rightarrow \infty} \psi_n(\lambda, t) = \text{exist} = \psi(\lambda, t)$

$$\frac{d}{dx} e^{A(x)} = \int_0^1 e^{(1-s)A(x)} A'(x) e^{sA(x)} ds = \int_0^1 e^{sA(x)} A'(x) e^{(1-s)A(x)} ds$$

15. Extensive property in stochastic models

$$\frac{\partial}{\partial t} P(\{\sigma_j\}, t) = \Gamma P(\{\sigma_j\}, t) ; \Gamma = \sum_j \Gamma_j$$

where  $\sigma_j = \pm 1$  (spin system) and

$$\Gamma_j P(\{\sigma_j\}, t) = -W_j(\sigma_j) P(\dots, \sigma_j, \dots) + W_j(-\sigma_j) P(\dots, -\sigma_j, \dots)$$

16. Theorem II (stochastic).

$$\text{If } P(st) \equiv e^{\text{st}(\Gamma_1 + \mu\Gamma_2)} e^{\mathcal{H}(i)} \in \mathcal{N}$$

("normal") for any  $\Gamma_1$  (and  $\Gamma_2 = \Gamma - \Gamma_1$ ) and for  $0 \leq s \leq 1$  and  $0 \leq \mu \leq 1$ , then

$$\lim_{\Omega \rightarrow \infty} \psi_{\Omega}(\lambda, t) = \text{exist} \quad (\text{uniformly convergent})$$

$$\Rightarrow \psi(\lambda, t) \text{ and } P(X, t) \text{ have extensive property.}$$

 17. Lemma 1. — If  $f(\{\sigma_j\}) \leq g(\{\sigma_j\})$ , then  $e^{t\Gamma} f \leq e^{t\Gamma} g$  for  $t \geq 0$ .

Lemma 2. — If  $f(\{\sigma_j\}) \in \mathcal{N}$  then  $|\Gamma_2 f| \leq C_{\Omega_2} f$ , where  $C_{\Omega_2}$  is a constant dependent on  $\Omega_2$ ,

$$\text{Def. } P \in \mathcal{N} ("normal") \Leftrightarrow P(\dots, -\sigma_j, \dots, t) \leq C_3 P(\dots, \sigma_j, \dots),$$

where  $C_3$  is a constant independent of  $\Omega$ ,

 18. Average motion, Fluctuation, and Nonlinear response

most probable path  $y(t)$  for  $\Omega$  large:

$$1) \quad x = y(t) + z, \quad g(z, t) \equiv \phi(y+z, t) = \{\psi(\lambda(y, t), t) - y\lambda(y, t)\} \\ - z \left\{ \lambda(y, t) + \left[ y - \left( \frac{\partial \psi}{\partial \lambda} \right)_{x=y} \right] \left( \frac{\partial \psi}{\partial \lambda} \right)_{x=y} \right\} + \dots,$$

$$\Rightarrow y(t) = \left( \frac{\partial \psi}{\partial \lambda} \right)_{\lambda=0} = \langle x \rangle_t; \quad \langle X \rangle_t = \int X \rho(X, t) dX = T_r X \rho(t).$$

$$2) \quad \text{variance: } \sigma(t) = \left( \frac{\partial^2 \psi}{\partial \lambda^2} \right)_{\lambda=0} = \Omega \langle (x - \langle x \rangle_t)^2 \rangle_t.$$

$$3) \quad \text{Nonlinear response: case (a); } \tilde{\mathcal{H}}^{(i)} = \mathcal{H}^{(i)} + K Y$$

$$y(t) = \langle x \rangle_{t,k}^{(a)} \equiv \Omega^{-1} \left( \frac{\partial \mathcal{F}}{\partial \lambda} \right)_{\lambda=0} = T_r x e^{\tilde{\mathcal{H}}^{(i)}(t)} / T_r e^{\mathcal{H}^{(i)}}$$

Theorem IIIa. — The nonlinear response for case (a) is

expressed by the fluctuation in non-equilibrium as

$$\Phi_{XY;Y}^{(a)} \equiv \frac{\partial y(t)}{\partial K} = \Omega^{-1} \int_0^1 d\mu \langle \delta X \delta Y(t; \mu) \rangle_{t,K}^{(a)} \equiv \Omega^{-1} \langle \delta X; \delta Y(t) \rangle_{t,K}^{(a)}$$

for a quantal system, where

$$Y(t; \mu) = e^{\mu \tilde{\mathcal{H}}^{(i)}} Y(t) e^{-\mu \tilde{\mathcal{H}}^{(i)}} \quad \text{and} \quad Y(t) = e^{-it\mathcal{H}} Y e^{it\mathcal{H}}.$$

For stochastic systems, we have

$$\Phi_{XY;Y}^{(a)} \equiv \frac{\partial Y(t)}{\partial K} = \Omega^{-1} \langle \delta X \delta Y(t) \rangle_{t,K}^{(a)};$$

$$Y(t) = (e^{t\Gamma} e^{\tilde{\mathcal{H}}^{(i)}})^{-1} e^{t\Gamma} (Y e^{\tilde{\mathcal{H}}^{(i)}}).$$

$$1) \quad \langle \delta X; \delta Y(t) \rangle_{t,K}^{(a)} = \langle \delta Y(t); \delta X \rangle_{t,K}^{(a)},$$

$$2) \quad \langle \delta X; \delta Y(t) \rangle_{t,K}^{(a)*} = \langle \delta Y(t); \delta X \rangle_{t,K}^{(a)},$$

$$3) \quad \Phi_{XY;Y}^{(a)} = \text{real}.$$

For a general nonlinear disturbance of the form  $\tilde{\mathcal{H}}^{(i)} = \mathcal{H}_K^{(i)}$ ,

$$\Phi_{XY;Y}^{(a)} \equiv \frac{\partial Y(t)}{\partial K} = \langle \delta X; \delta \left( \frac{\partial}{\partial K} \mathcal{H}_K^{(i)}(t) \right) \rangle_{t,K}^{(a)}.$$

case (b)  $\tilde{\mathcal{H}} = \mathcal{H} + KY$ :

$$y(t) = \langle x \rangle_{t,K}^{(b)} \equiv \Omega^{-1} \left( \frac{\partial \mathcal{Z}}{\partial \lambda} \right)_{\lambda=0} = \text{Tr } x e^{-it\tilde{\mathcal{H}}} e^{\mathcal{H}^{(i)}} e^{it\tilde{\mathcal{H}}}$$

Theorem IIIb.

$$\Phi_{XY;Y}^{(b)} \equiv \frac{\partial y(t)}{\partial K} = -\frac{i}{\Omega} \int_0^t \langle [\mathbf{X}, \mathbf{Y}(s)] \rangle_{t,K}^{(b)} ds,$$

for a quantal system where  $Y(s) = \exp(-is\tilde{\mathcal{H}}) Y \exp(is\tilde{\mathcal{H}})$ .

$$\Phi_{XY;Y}^{(b)} = \sum x e^{t\Gamma_K} \int_0^t ds e^{-s\Gamma_K} \left( \frac{d}{dK} \Gamma_K \right) e^{s\Gamma_K} e^{\mathcal{H}^{(i)}}$$

for a stochastic system, where  $\Gamma_K \leftrightarrow \tilde{\mathcal{H}} = \mathcal{H} + KY$ .

For a general nonlinear disturbance of the form  $\tilde{\mathcal{H}} = \mathcal{H}_K$ ,

$$\Phi_{XY;Y}^{(b)} \equiv \frac{\partial}{\partial K} y(t) = -i \int_0^t \langle [x, \left( \frac{\partial}{\partial K} \mathcal{H}_K \right)(s)] \rangle_{t,K}^{(b)} ds.$$

## 19. Relation between $\phi(x, t)$ and cumulants

Theorem IV. — If  $\Psi(\lambda, t)$  (and  $\rho(x, t)$ ) has extensive property,



$\langle \mathbf{X}^n \rangle_{c,t}$  = extensive (i.e.,  $O(\Omega)$ ). Conversely, if  $\langle \mathbf{X}^n \rangle_{c,t}$  are extensive, and

if  $\lim_{n \rightarrow \infty} (\langle \mathbf{X}^n \rangle_{c,t} / n!)^{1/n} = A_0^{-1} < \infty$ , then  $\Psi(\lambda, t)$  and  $\rho(x, t)$  exist and have extensive property.

20. Theorem V. — The function  $\phi(x, t)$  is expressed by the cumulants

$$\text{as } \phi(y+z, t) = - \sum_{n=2}^{\infty} \frac{z^n}{n!} \lim_{\Omega \rightarrow \infty} f_n(\langle \mathbf{X}^2 \rangle_{c,t} \Omega^{-1}, \dots, \langle \mathbf{X}^n \rangle_{c,t} \Omega^{-1}),$$

where  $f_n$  is defined by

$$(\psi_2^{-1} \partial / \partial \lambda)^{n-2} \psi_2^{-1} \equiv f_n(\psi_2, \psi_3, \dots, \psi_n); \psi_n \equiv \partial^n \psi / \partial \lambda^n,$$

and  $\psi_2^{2n-3} f_n$  is a homogeneous polynomial of order  $(n-2)$ .

21. Explicit forms of  $\{f_n\}$ :

$$f_2 = \psi_2^{-1}, f_3 = -\psi_3 \psi_2^{-3}, f_4 = -(\psi_2 \psi_4 - 3\psi_2^2) \psi_2^{-5}, \dots$$

22. Theorem VI. When  $\phi(y+z, t) = \sum_{n=2}^{\infty} a_n z^n / n!$  is known, all

cumulants  $\langle \mathbf{X}^n \rangle_{c,t}$  are obtained explicitly with use of the recursion formula;

$$\begin{aligned} \langle \mathbf{X}^n \rangle_{c,t} = \Omega \{ a_n [\sigma(t)]^n + \\ + [\sigma(t)]^{3-n} g_n(\langle \mathbf{X}^2 \rangle_{c,t} \Omega^{-1}, \dots, \langle \mathbf{X}^{n-1} \rangle_{c,t} \Omega^{-1}) \}, \end{aligned}$$

for  $n \geq 3$ , and for large  $\Omega$ , where  $g_n$  is given by

$$g_n(\psi_2, \dots, \psi_{n-1}) = -\psi_2^{2n-3} f_n - \psi_2^{n-3} \psi_n.$$

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23. Explicit forms of  $\langle \mathbf{X}^n \rangle_{c,t}$  are

$$\langle \mathbf{X}^3 \rangle_{c,t} = \Omega a_3 [\sigma(t)]^3, \langle \mathbf{X}^4 \rangle_{c,t} = \Omega \{ a_4 [\sigma(t)]^4 + 3a_3^2 [\sigma(t)]^5 \}, \dots$$

24. Composed macrovariable  $f(x)$  ;  $x = \mathbf{X}/\Omega$  :

$$\langle f(x) \rangle = f(y(t)) + O(\Omega^{-1}),$$

and the variance  $\sigma_f(t)$  is given by

$$\sigma_f(t) = \left( \frac{\partial f}{\partial x} \right)_{x=y(t)}^2 \sigma_x(t) + O(\Omega^{-1}),$$

25. Exactly soluble models:

Ex.1 Non-interacting system:

$$\mathcal{H}_{\text{NI}} = -J \sum_j \sigma_j^x, \quad X = \sum_{j=1}^{\Omega} \sigma_j^z \quad \text{and} \quad \mathcal{H}^{(i)} = h \sum_{j=1}^{\Omega} \sigma_j^z.$$

Solution:  $\Psi(\lambda, t) = \exp [\Omega \psi(\lambda, t)]$  (exact for any  $\Omega$ ),

$$\psi(\lambda, t) = \log \{ \cosh \lambda + \sinh \lambda \tanh h \cos(2tJ/h) \},$$

$$\lambda(x, t) = \tanh^{-1} \{ [x - a(t)] / [1 - a(t)x] \}; \quad a(t) = \tanh h \cos(2tJ/h),$$

$$\phi(x, t) = \log \{ a(t) \sinh \lambda(x, t) + \cosh \lambda(x, t) \} - x \lambda(x, t)$$

$$y(t) = a(t) \quad \text{and variance} \quad \sigma(t) = 1 - y^2(t)$$

Ex.2 Kinetic Weiss Ising model (Kubo et al, .....

Ex.3 (Anisotropic) Weiss Heisenberg model

$$\mathcal{H} = \mathcal{H}(J_x, J_y, J_z; H) = -\Omega^{-1} \sum_{i,j} (J_x \sigma_i^x \sigma_j^x + J_y \sigma_i^y \sigma_j^y + J_z \sigma_i^z \sigma_j^z) - \mu H \sum_j \sigma_j^z,$$

$$X = \sum_j \sigma_j^z, \quad \text{and} \quad \mathcal{H}^{(i)} = -\beta \mathcal{H}(J_x^0, J_y^0, J_z^0; H_0)$$

Ex.4 Nonlinear relaxation in the linear kinetic Ising model.

Ex.5 Nonlinear relaxation in the linear anisotropic XY-model:

$$\mathcal{H} = \mathcal{H}(J_x, J_y; H) = -\sum_j^{\Omega} (J_x \sigma_j^x \sigma_{j+1}^x + J_y \sigma_j^y \sigma_{j+1}^y) - \mu H \sum_j \sigma_j^z,$$

$$X = \sum_j \sigma_j^z \quad (\text{or } \sum_j \sigma_j^x \sigma_{j+1}^x \text{ etc.}), \quad \mathcal{H}^{(i)} = -\beta \mathcal{H}(J_x^0, J_y^0; H^0).$$

## 26. Applications: relaxation and fluctuation in superconductors.

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